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Optical properties of tellurium-doped $In_xGa_{1-x}As_ySb_{1-y}$ epitaxial layers studied by photoluminescence spectroscopy

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Abstract
Controlled doping of quaternary alloys of $In_xGa_{1-x}As_ySb_{1-y}$ with tellurium is fundamental to obtain the n-type layers needed for the development of optoelectronic devices based on p–n heterojunctions. InGaAsSb epitaxial layers were grown by liquid phase epitaxy and Te doping was obtained by incorporating small Sb_3Te_2 pellets in the growth melt. The tellurium doping levels were in the range 10^{16}–10^{17} cm^{-3}. We have used low-temperature photoluminescence (PL) spectroscopy to study the influence of the Te donor levels on the radiative transitions shown in the PL spectra. The PL measurements were done by exciting the samples with the 448 nm line of an Ar ion laser with varying excitation powers in the range from 10 to 200 mW. For the low-doped sample the PL spectrum showed a narrow exciton-related peak centred at around 610 meV with a full width at half maximum (FWHM) of about 7 meV which is evidence of the good crystalline quality of the layers. For higher Te doping, the PL spectra show the presence of band-to-band and donor-to-acceptor transitions which overlap as the Te concentration increases. The peak of the PL band shifts to higher energies as Te doping increases due to a band-filling effect as the Fermi level enters into the conduction band. From the peak energy of the PL spectra, and using a model that includes the band-filling and band-shrinkage effects due to the carriers, we have estimated the effective carrier concentration due to doping with Te in the epilayers.

1. Introduction

During the last two decades much time has been devoted to the study of narrow-gap semiconductors due to the development of optoelectronic devices in the infrared range, with

important applications in different areas of science and technology. The $In_xGa_{1-x}As_ySb_{1-y}$ alloy system is interesting for optoelectronic applications because its room-temperature band-gap energies cover an extremely wide range from 0.10 to 1.42 eV. For InGaAsSb layers lattice-matched to InP substrates, the band-gap energy variation is nearly fixed, ranging from 0.74 to 0.78 eV. However, for InGaAsSb layers lattice-matched to GaSb, the band-gap energy varies between 0.29 and 0.73 eV, corresponding to the wavelength range from 1.7 to 4.3 μm. This is a very interesting wavelength range for anticipated future fibre optics communication systems. The development of new fibres, such as the heavy metal fluoride fibres, offers the possibility of lowering the losses for these fibres by one or two orders of magnitude compared with conventional SiO_2 fibres; the minimum loss is expected to occur in the wavelength range from 2 to 4 μm [1, 2]. The quaternary alloy $In_xGa_{1-x}As_ySb_{1-y}$ lattice-matched to GaSb is a narrow-band-gap semiconductor (0.3–0.7 eV) with a great number of applications including thermophotovoltaic cells [3], infrared light-emitting diodes and lasers, and photodetectors [4].

The presence of a large background of defects or impurities in GaSb and its alloys leads to numerous acceptor-bounded exciton luminescence bands. Many of these transitions appear to be growth specific. The PL spectra of the melt-grown Te-doped GaSb single crystals have been extensively studied. Tellurium is known to form a shallow donor level; in addition, it has been reported to introduce deep levels [5] which behave as acceptor-like and are close in energy to the native residual acceptors in undoped GaSb and its alloys. This has led to some confusion in the identification of these levels. Recombination transitions involving neutral residual acceptors, ionized residual acceptors, deep acceptors and some other deep acceptor levels have been reported [6, 7]. The dominance of any of these transitions was found to depend on the Te doping level and the incident excitation intensity. There are few reports on luminescence studies for liquid phase epitaxy (LPE)-grown Te-doped InGaAsSb epilayers. A Burstein–Moss shift has been reported in Te-doped InGaAsSb epilayers as well as broadening of the spectra with an increase in the carrier concentration, but the investigated doped range was up to 10^{17} cm^{-3} [6].

In this work we have carried out a systematic study and quantitative evaluation of the effects of Te impurification in InGaAsSb alloys as a function of Te doping using low-temperature (15 K) photoluminescence (PL) spectroscopy for a set of samples where the Te doping was varied in a broad range: from low doping levels using GaSb:Te to dope the epilayers, up to higher doping levels reached by using pieces of Sb_3Te_2 for doping. The PL spectra showed a broadening of the main emission band as the Te concentration increased, and a shift to higher energies of the PL peak. We have interpreted these results in terms of combined band filling and band shrinkage effects; and from the shift of the Fermi level deep into the conduction band, we have estimated the resulting electron concentration for the different samples.

2. Experimental details

$In_xGa_{1-x}As_ySb_{1-y}$ layers were grown by liquid phase epitaxy (LPE) on Te-doped (100) GaSb substrates under lattice-matching conditions. Details of the growth conditions have been published elsewhere [8]. Low Te doping was obtained by adding Te-doped GaSb pieces to the growth melt (sample 105) and high Te doping was accomplished by adding Sb_3Te_2 pellets to the growth melt; three samples, 102, 103 and 108 were grown by adding 0.15, 0.32 and 0.63 mg of Sb_3Te_2 respectively. PL measurements were carried out by exciting the sample with the 488 nm line of an Ar-ion laser and varying the exciting power in the range between 40 and 200 mW. Sample radiative emission was analysed through an Acton monochromator and detected with an EG&G Judson InSb infrared detector cooled with liquid nitrogen. The chemical stoichiometry of the epitaxial layers is roughly $In_{0.14}Ga_{0.86}As_{0.13}Sb_{0.87}$ and was

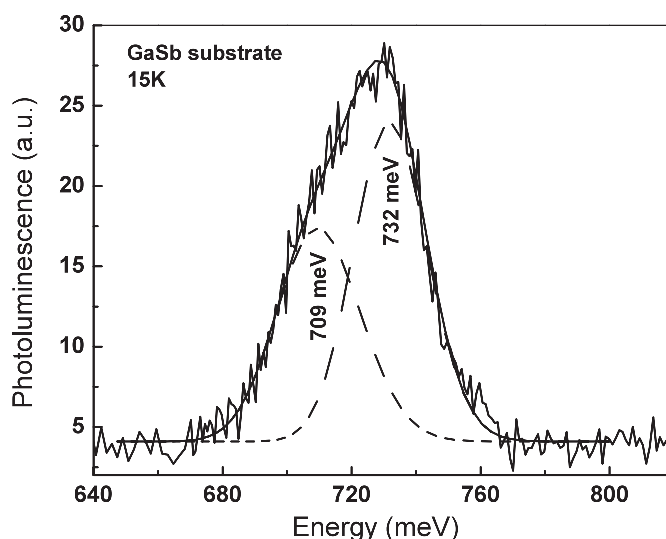

Figure 1. Photoluminescence spectrum for the Te-doped (100)GaSb substrate at 15 K. The solid lines correspond to the Gaussian-like fitting.

determined by energy dispersive spectroscopy (EDS)-scanning electronic microscopy [8]. The accuracy in the determination of x and y was about 2%.

The energy positions and the full width at half maximum (FWHM) of each peak in the PL spectra have been determined by a quantitative fit to the experimental PL spectra using a sum of Gaussian-like distributions, the dominant peaks were fitted first and the additional peaks were added as necessary.

3. Results and discussion

In figure 1 we show a typical low-temperature PL spectrum of the GaSb substrate used in this work. As can be observed in the figure, the spectrum exhibits an asymmetric PL emission band centred at 727 meV with a FWHM of about 46 meV. We have fitted this experimental PL band with two Gaussian bands to account for the shoulder observed at the low-energy side of the peak; from this fitting process we obtained that this band is constituted of two radiative transitions peaking at 709 and 732 meV respectively. The peak sited at 732 meV is attributed to a transition from the conduction band to an acceptor consisting of a Te–Ga antisite complex of the type $V_{Ga}Ga_{Sb}Te_{Sb}$ lattice defect [9]. The transition at 709 meV is coincidental with the second ionization level of the residual acceptor [10, 11, 7].

In figure 2 we show the PL spectra for an epitaxial layer with a low Te concentration ($In_{0.14}Ga_{0.86}As_{0.13}Sb_{0.87}$; sample 105) measured at 15 K. The PL spectrum is typical for samples with a low free carrier concentration. This spectrum presents two sharp emission bands centred at 648.6 and 614 meV (labelled as BE and A [12]) with FWHMs of 7 and 18 meV, respectively; and also a weak broad band at 560 meV. These bands do not change noticeably with increasing laser power excitation, as can be observed in figure 2. We suggest that the transition labelled as BE is associated with the decay of an exciton bound to a neutral acceptor. Our assignment is primarily based on the value of its linewidth [12]. Thus, if we assume that BE is an acceptor–bound exciton transition, we can estimate the band-gap energy for this $In_{0.14}Ga_{0.86}As_{0.13}Sb_{0.87}$ alloy by adding approximately 4 meV (namely, the sum of the exciton dissociation energy plus

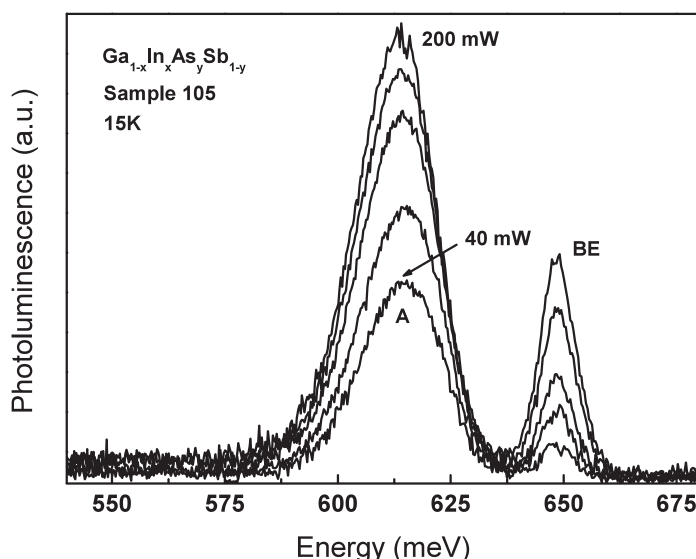

Figure 2. Low-temperature PL spectra for $In_{0.14}Ga_{0.86}As_{0.13}Sb_{0.87}$ epilayers for low Te concentration. Experimental spectra correspond to different excitation laser powers for sample 105.

the free exciton (FE) binding energy) to the transition energy of the BE, namely 648.6 meV. This gives us a band-gap energy of about 653 meV for this epitaxial layer. In addition to the above transition, we also observe a transition corresponding to the free electron-to-acceptor recombination (A) sited at 614 meV, being the dominant transition. In GaSb and its alloys the donor levels are shallow (∼2–3 meV) due to the small effective mass of the electron, which makes it difficult to distinguish between free-to-bound and excitonic recombination processes. From the value of the estimated band-gap energy, and the energy of transition A we determine the binding energy of the acceptor to be 39 meV. This value compares quite well with those determined in GaSb (34 meV) and GaInAsSb (38 meV) by Iyer *et al* [12].

To further confirm the identity of the various emission bands of sample 105, the PL spectra were studied as a function of the incident power intensity. The integrated intensity of the BE transition as a function of the exciting laser power for sample 105 is depicted in figure 3. It exhibits a power-law dependence of the type $P^{1.3}$. The transition identified as A exhibits a power-law dependence of approximately $P^{0.5}$. These values are somewhat different than the power-law dependence reported for GaSb layers ($P^{0.9}$) [5] and those reported by Iyer *et al* [12] for InGaAsSb layers ($P^{1.0}$–$P^{2.0}$), where they found that the power-law parameter was dependent on various recombination parameters which were highly sample dependent. Also, it is important to notice that no saturation effect in any of the PL peaks was observed in the limited range of incident laser powers used for this sample.

As an important part of this work, we have made a systematic study of doping of InGaAsSb epilayers with tellurium at higher concentrations. Tellurium has been reported to be a shallow n-type impurity for GaSb and its alloys. In figure 4 we present the PL spectra for the complete set of Te-doped samples for a laser excitation power of 200 mW. As compared to the PL spectrum for the low-doped sample, the PL spectra for the higher-doped samples present a broadening of the band corresponding to the band-to-band transition (BB) that overlaps with the band-to-acceptor transition (A), and the appearance of a broad low-energy band (in the range 550–600 meV). Both of these bands become broader and shift to higher energies as the Te

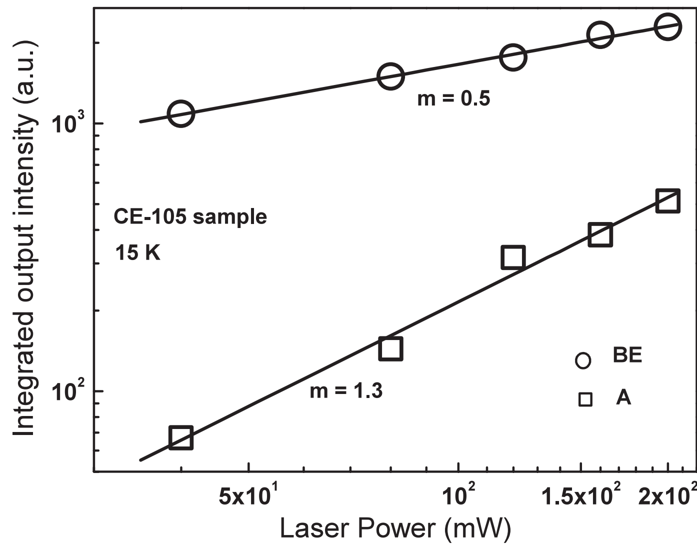

Figure 3. Dependence on the power excitation of the integrated PL intensities of the 648.6 meV band (BE, open circles) and the 614.0 meV band (A, open squares). Solid lines are linear regressions to the points.

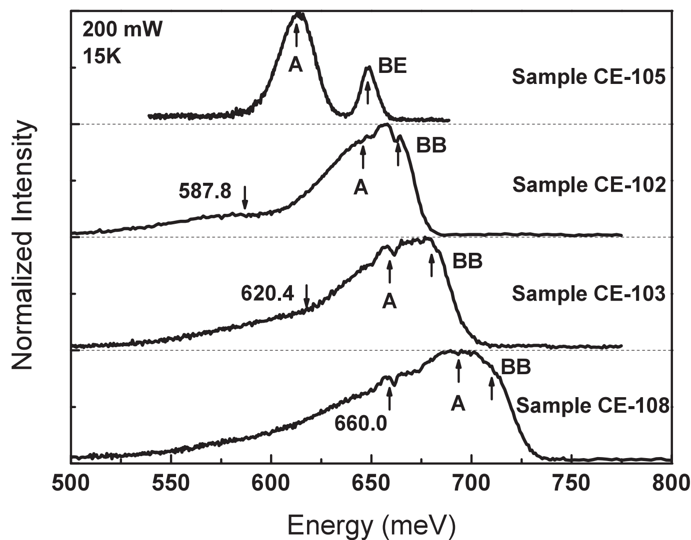

Figure 4. PL spectra for $In_{0.14}Ga_{0.86}As_{0.13}Sb_{0.87}$:Te layers, as a function of the Sb_3Te_2 content in the growth melt, measured at 15 K, and for 200 mW of excitation power.

concentration in the InGaAsSb layer increases. Additionally, at very high concentrations the luminescence band becomes highly asymmetric, which is particularly striking at the excitation intensities studied [13].

In order to account for the peak position and the half-width of the PL band, it is necessary to include not only the band-filling effect due to the high carrier density, but also the band-gap shrinkage due to the exchange interaction between free carriers. In the heavily doped n-GaInAsSb epilayers, the quasi-Fermi level for electrons moves into the conduction band;

and so, the band-to-band absorption process takes place for energies larger than the band gap energy, and between states with $k > 0$.

The density of electrons in the conduction band, assuming a parabolic density of states and assuming also that the electron effective mass does not depend on the concentration (although this assumption is inadequate for high concentrations [14]), takes the form:

$$n = 4\pi \left(\frac{2m_n^*}{h^2}\right)^{3/2} (kT)^{3/2} \int_0^\infty \frac{\varepsilon^{1/2}\,d\varepsilon}{e^{\varepsilon-\eta+1}+1} = N_c F_{1/2}(\eta) \qquad (1)$$

where η is the reduced Fermi level, $\eta = (F - E_c)$ and $\varepsilon = (E - E_c)$. N_c is the effective density of states in the conduction band, and $F_{1/2}(\eta)$ is the Fermi–Dirac integral of grade 1/2, and they are given by:

$$N_c = 2\left(\frac{2\pi m_n^* kT}{h^2}\right)^{3/2} \qquad (2)$$

$$F_{1/2}(\eta) = \frac{2}{\sqrt{\pi}} \int_0^\infty \frac{\varepsilon^{1/2}\,d\varepsilon}{e^{\varepsilon-\eta+1}+1}. \qquad (3)$$

A semiconductor is considered completely degenerated if $\eta > 5$; that is, when the Fermi level is situated at least to $5kT$ over E_c. In a heavily doped n-type semiconductor, the Fermi level moves significantly into the conduction band, and at low temperatures all electron states below the Fermi level are occupied, making the threshold energy greater than energy gap E_g by an amount which equals the Fermi level [15]. The electron concentration n of a heavily doped n-type semiconductor as a function of the Fermi level $(F - E_c)$ is expressed as follows [15]:

$$n = \frac{8\pi}{3}\left(\frac{2m_n^*}{h^2}\right)^{3/2} (F - E_c)^{3/2}. \qquad (4)$$

From the above equation it is deduced that the free carrier concentration in a completely degenerate semiconductor does not depend on the temperature; it is determined by the position of the Fermi level, F, and the magnitude of the carrier effective mass.

For high doping levels, the band-gap shrinkage due to the electron–electron interaction should also be taken into account since it influences the optical properties of the semiconductor [16, 17], and consequently, the properties of the optoelectronic device [18, 19]. According to Camassel [20], this shrinkage depends on the electron concentration as:

$$E_c^e = (2e^2/\varepsilon_\tau)(3/\pi)^{1/3} n^{1/3} \qquad (5)$$

so its strength depends essentially on the dielectric constant of the material and the electron concentration. Experimental observations of the band-gap shrinkage have been observed in GaAs [21], with a proportionality constant of around 1.6×10^{-8}, and in InP [15], with a constant of 2.25×10^{-8}; these value constants are expressed for E_c^e in eV, and for n in cm^{-3}.

In this way, taking into account the band-filling effect and the band-gap shrinkage, we will have the following relationship between the Fermi level and the electron concentration:

$$n^{2/3} = A^{2/3}(F - E_{g0} + Bn^{1/3}). \qquad (6)$$

where

$$A = \left(\frac{8\pi}{3}\right)\left(\frac{2m_n^*}{h^2}\right)^{3/2}; \qquad B = (2e^2/\varepsilon_\tau)(3/\pi)^{1/3}. \qquad (7)$$

In our case, if we take the electron effective mass, m_n^*, as $0.0354\, m_0$ for n-type GaInAsSb, and considering that the peak labelled as BB in figure 4 corresponds to the values of the Fermi level, F, we can estimate the free electron concentration in the Te-doped InGaAsSb epilayers. The results are presented in the table 1, where we observe that the carrier concentrations determined

Table 1. Electron concentrations obtained from the PL spectra.

Sample	% Te in the growth melt	$F - E_{g0}$ (meV)	η	n (cm^{-3})
105	0.000 42	—	—	—
102	0.006 3	7	5.38	5.3×10^{16}
103	0.013 4	23	17.69	2×10^{17}
108	0.026 3	38	29.23	3.6×10^{17}

from equation (6) and the PL spectra follow roughly the proportional amount of Sb_3Te_2 used in the growth melt, even when the level of doping is lower than expected from the amount of Te present in the melt. We believe that a possible explanation is related to a compensation effect where some of the Te donors are neutralized by the Sb-acceptor vacancies which are always present in the growth of this type of quaternary layer.

4. Conclusions

To summarize: in this work we have presented a systematic study of doping by tellurium in $In_{0.14}Ga_{0.86}As_{0.13}Sb_{0.87}$ epitaxial layers grown on (100)GaSb substrates by LPE. The epilayers studied were doped with Te in a broad range from low to high concentrations. For low doping levels, the PL spectra show the presence of exciton-related transitions with a small FWHM value (7 meV), which is evidence of good crystalline quality of the layer. For higher doping levels, the PL spectra exhibit band-to-band and donor-to-acceptor transitions which merge in a broad band as the Te doping increases. From the peak of the PL spectra, and using a model that takes into account the band-filling and band-shrinkage effects, we were able to determine the effective carrier concentration due to the Te doping which was smaller than expected, probably due to a compensation effect between the acceptor-like Sb vacancies and the donor-like Te impurities.

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